

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

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P-01,0300

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)

09/937290

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PRIORITY DATE CLAIMED

PCT/DE00/00783

March 13, 2000

March 24, 1999

TITLE OF INVENTION "ORGANIC ELECTROLUMINESCENT COMPONENT"

APPLICANT(S) FOR DO/EO/US

Andreas Kanitz and Matthias Stössel

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay.
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. §371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98; (PTO 1449, Prior Art, Search Report).
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☒ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:
 - a. ☒ Submission of Drawings with Translations of German-Language Legends - 3 sheets
 - b. ☒ EXPRESS MAIL #EL843743118US dated September 24, 2001

09/937290

17. ☒ The following fees are submitted:**BASIC NATIONAL FEE (37 C.F.R. 1.492(a)(1)-(5):**

Search Report has been prepared by the EPO or JPO \$860.00

International preliminary examination fee paid to USPTO (37 C.F.R. 1.482) ... \$670.00

No international preliminary examination fee paid to USPTO (37 C.F.R. 1.482) but
international search fee paid to USPTO (37 C.F.R. 1.445(a)(2)) \$760.00Neither international preliminary examination fee (37 C.F.R. 1.482) nor international
search fee (37 C.F.R. 1.445(a)(2)) paid to USPTO \$970.00International preliminary examination fee paid to USPTO (37 C.F.R. 1.482) and all
claims satisfied provisions of PCT Article 33(2)-(4) \$ 96.00**ENTER APPROPRIATE BASIC FEE AMOUNT =**

CALCULATIONS

PTO USE ONLY

\$ 860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from
the earliest claimed priority date (37 C.F.R. 1.492(e)).

\$

Claims

Number Filed

Number
Extra

Rate

Total Claims

18

- 20 =

0

X \$18.00

\$

Independent Claims

1

- 3 =

0

X \$80.00

\$

Multiple Dependent Claims

\$270.00 +

\$

TOTAL OF ABOVE CALCULATIONS =

\$ 860.00

Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also
be filed. (Note 37 C.F.R. 1.9, 1.27, 1.28)

\$

SUBTOTAL =

\$ 860.00

Processing fee of \$130.00 for furnishing the English translation later than ☐ 20 ☐ 30 months
from the earliest claimed priority date (37 CFR 1.492(f)).

+

\$

TOTAL NATIONAL FEE =

\$ 860.00

Fee for recording the enclosed assignment (37 C.F.R. 1.21(h). The assignment must be
accompanied by an appropriate cover sheet (37 C.F.R. 3.28, 3.31). \$40.00 per property

+

TOTAL FEES ENCLOSED =

\$ 860.00

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refunded

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- a. ☒ A check in the amount of \$860.00 to cover the above fees is enclosed.
- b. ☐ Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 501519. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 C.F.R. 1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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SIGNATURE

James D. Hobart

NAME

24,149

Registration Number

- 1 -

IN THE UNITED STATES ELECTED OFFICE OF
THE UNITED STATES PATENT AND TRADEMARK OFFICE
UNDER THE PATENT COOPERATION TREATY - CHAPTER II

SUBMISSION OF DRAWINGS WITH
TRANSLATIONS OF GERMAN-LANGUAGE LEGENDS

APPLICANTS: Andreas Kanitz and Matthias Stössel

ATTORNEY

DOCKET NO.: P-01,0300

SERIAL NO.:

EXAMINER:

FILING DATE:

ART UNIT:

INTERNATIONAL APPLICATION NO.: PCT/DE00/00783

INTERNATIONAL FILING DATE: 13 March 2000

INVENTION: "ORGANIC ELECTROLUMINESCENT COMPONENT"

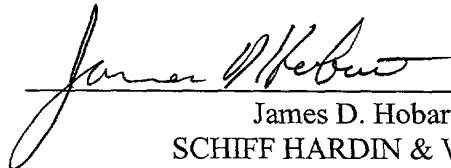
BOX PCT

Assistant Commissioner for Patents
Washington, D.C. 20231

S I R:

Attached herewith are three sheets of drawings containing Figs. 1-5, with
the legends on the graphs of Figs. 3, 4 and 5 being provided with translations.

Respectfully submitted,

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DATED: September 24, 2001

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IN THE UNITED STATES ELECTED OFFICE OF
THE UNITED STATES PATENT AND TRADEMARK OFFICE
UNDER THE PATENT COOPERATION TREATY - CHAPTER II

PRELIMINARY AMENDMENT

5 APPLICANTS: Andreas Kanitz and Matthias Stössel

ATTORNEY

DOCKET NO.: P-01,0300

SERIAL NO.:

EXAMINER:

FILING DATE:

ART UNIT:

10 INTERNATIONAL APPLICATION NO.: PCT/DE00/00783

INTERNATIONAL FILING DATE: 13 March 2000

INVENTION: "ORGANIC ELECTROLUMINESCENT COMPONENT"

BOX PCT

15 Assistant Commissioner for Patents
Washington, D.C. 20231

S I R:

Please amend the above-identified International Application before entry
into the National Stage before the U.S. Patent and Trademark Office under 35 USC
371 as follows:

20 IN THE SPECIFICATION:

Please replace pages 1-15 with the attached Substitute Specification.

IN THE ABSTRACT OF THE DISCLOSURE:

Please replace the Abstract with the attached unnumbered page containing
the Abstract of the Disclosure.

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- 2 -

IN THE CLAIMS:

Please cancel claims 1-10, without prejudice.

Please add the following claims.

5 --11. An organic electroluminescent component for an organic light-emitting diode, said component comprising:

a transparent bottom electrode situated on a substrate;

a top electrode composed of a metal that is inert to oxygen and moisture;

at least one organic function layer arranged between the bottom electrode and the top electrode; and

10 a charge carrier injection layer containing a complex metal salt of the composition $(Me1)(Me2)F_{m+n}$, whereby the following applies:

m and n are respectively a whole number corresponding to the valence of the metals Me1 and Me2 with the metal Me1 having the valence m, the metal Me2 having the valence n,

15 Me1 being a metal selected from a group consisting of Li, Na, K, Mg and Ca,

Me2 being a metal selected from a group consisting of Mg, Al, Ca, Zn, Ag, Sb, Ba, Sm and Yb,

with the prescription: $Me1 \neq Me2$.--

20 --12. An organic electroluminescent component according to claim 11, wherein the top electrode is composed of a metal selected from a group consisting of aluminum, silver, platinum and gold and of an alloy of two of these metals.--

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--13. An organic electroluminescent component according to claim 12, wherein the charge carrier injection layer is arranged between the top electrode and the uppermost organic function layer.--

5 --14. An organic electroluminescent component according to claim 13, wherein the charge carrier injection layer comprises a thickness between 0.1nm and 20nm.--

--15. An organic electroluminescent component according to claim 14, wherein the metal Me1 is lithium and the metal Me2 is selected from magnesium, aluminum, calcium, silver and barium.--

10 --16. An organic electroluminescent component according to claim 15, wherein the complex metal salt is a salt selected from LiAlF_4 , LiAgF_2 and LiBaF_3 .--

15 --17. An organic electroluminescent component according to claim 16, wherein two organic function layers are arranged between the bottom electrode and the top electrode, wherein an apertured conducting layer is located on the bottom electrode and an emission layer is located on said conducting layer.--

20 --18. An organic electroluminescent component according to claim 17, wherein the apertured conducting layer contains a material selected from N,N'-bis-(3-methylphenyl)-N,N'-bis(phenyl)-benzidine; 4,4',4''-tris-(N-1-naphthyl-N-phenylamino)-triphenylamine; and N,N'-bis-phenyl-N,N'-bis- α -naphthyl-benzidine and the emission layer is a hydroxyquinoline aluminum-III salt.--

--19. An organic electroluminescent component according to claim 18, wherein the bottom electrode is composed of indium tin oxide.--

--20. An organic electroluminescent component according to claim 19, wherein an electron transport layer is arranged on the at least one organic function layer.--

5 --21. An organic electroluminescent component according to claim 11, wherein the charge carrier injection layer is arranged between the top electrode and an uppermost organic function layer.--

--22. An organic electroluminescent component according to claim 11, wherein the charge carrier injection layer comprises a thickness between 0.1nm and 20nm.--

10 --23. An organic electroluminescent component according to claim 11, wherein the metal Me1 is lithium and the metal Me2 is selected from magnesium, aluminum, calcium, silver and barium.--

--24. An organic electroluminescent component according to claim 23, wherein the complex metal salt is selected from LiAlF_4 , LiAgF_2 and LiBaF_3 .--

15 --25. An organic electroluminescent component according to claim 11, wherein two organic function layers are arranged between the bottom electrode and the top electrode, whereby an apertured conducting layer is located on the bottom electrode and an emission layer is located on said conducting layer.--

20 --26. An organic electroluminescent component according to claim 25, wherein the apertured conducting layer is a material selected from N,N'-bis-(3-methylphenyl)-N,N'-bis(phenyl)-benzidine; 4,4',4''-tris-(N-1-naphthyl-N-

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-27. An organic electroluminescent component according to claim 11,
wherein the bottom electrode is composed of indium tin oxide.--

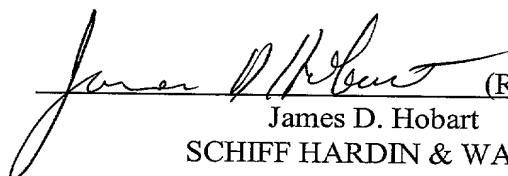
5 --28. An organic electroluminescent component according to claim 11,
wherein an electron transport layer is arranged on the at least one organic function
layer.--

REMARKS

Claims 11-28 are presented for examination.

By this amendment, the translation of the PCT Application has been amended, with the amendments being incorporated in the attached Substitute Specification. A marked-up copy of the translation of the specification showing changes made is also attached. Also attached herewith on an unnumbered page is a new Abstract of the Disclosure. A marked-up copy of the Abstract is also attached. Claims 1-10 have been cancelled and new claims 11-28 have been submitted, which have been carefully drafted to remove multiple-dependency and to be in U.S. form.

Respectfully submitted,

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DATED: September 24, 2001

ABSTRACT OF THE DISCLOSURE

The organic electroluminescent component of the invention has a transparent bottom electrode situated on a substrate; a top electrode composed of a metal that is inert to oxygen and moisture; at least one organic function layer
5 arranged between the bottom electrode and the top electrode; and a charge carrier injection layer containing a complex metal salt of the composition $(\text{Me1})(\text{Me2})\text{F}_{m+n}$, whereby the following applies:

m and n are respectively a whole number corresponding to the valence of the metals Me1 and Me2 (the metal Me1 thereby has
10 the valence m, the metal Me2 the valence n),

Me1 is selected from a group consisting of Li, Na, K, Mg and Ca,

Me2 is selected from a group consisting of Mg, Al, Ca, Zn, Ag, Sb, Ba, Sm and Yb,

with the prescription: $\text{Me1} \neq \text{Me2}$.

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Appendix

Title

1

ORGANIC ELECTROLUMINESCENT COMPONENT

Background of the Invention

The invention is directed to an organic electroluminescent component, particularly an organic light-emitting diode.

The visualization of data is constantly increasing in significance due to the great increase in the amount of information. The technology of flat picture screens ("flat panel displays") was developed therefor for employment in mobile and portable electronic devices. The market of flat panel displays is currently largely dominated by the technology of liquid crystal displays (LC displays). In addition to cost-beneficial manufacture, low electrical power consumption, low weight and slight space requirement, however, the technology of LC displays also exhibits serious disadvantages.

LC displays are not self-emitting and can therefore only be easily read or recognized given especially beneficial ambient light conditions. This makes a back-illumination device necessary in most instances, but this multiplies the thickness of the flat panel display. Moreover, the majority part of the electrical power consumption ^{of the display} is then needed for the illumination, and a higher voltage is required for the operation of the lamps or fluorescent tubes, ^{which higher voltage} this is usually generated from batteries or accumulators with the assistance of "voltage-up converters". Other disadvantages are the highly limited observation angles of LC displays and the long switching times of individual pixels, ^{which switching times typically lie} these typically lying at a few milliseconds and also being highly temperature-dependent. The delayed image build-up is considered extremely disturbing, for example, given utilization in means of conveyance or given video applications.

There are other flat panel display technologies in addition to LC displays, for example the technology of flat display panel cathode ray tubes, of vacuum-fluorescence displays and of inorganic thin-film electroluminescent displays. ^{However, either} ~~these~~ ^{there} technologies ~~however~~ have ~~either~~ not yet achieved the required degree of technological maturity or -- due to high operating voltages or, respectively, high

manufacturing costs -- they are only conditionally suited for utilization in portable electronic devices.

Displays on the basis of organic light-emitting diodes ^{which are called LEDs,} ~~(=OLEDs)~~ do not exhibit ^{these} ~~said~~ disadvantages. The necessity of a back-illumination is eliminated due to the self-emissivity, as a result whereof the space requirement and the electrical power consumption are considerably reduced. The switching times lie at about one microsecond and are only slightly temperature-dependent, which enables employment for video applications. The reading angle amounts to nearly 180°, and polarization films ^{that are} ~~as~~ required given LC displays are eliminated, so that a greater brightness of the display elements can be achieved. Further advantages are the employability of flexible and non-planar substrates as well as a simple and cost-beneficial manufacture.

The construction of organic light-emitting diodes typically ensues in the following way.

A transparent substrate, for example glass, is coated with a transparent electrode (bottom electrode, anode), composed, for example, of indium tin oxide (ITO). Dependent on the application, the transparent electrode is then structured with the assistance of a photolithographic process.

One or more organic layers composed of polymers, oligomers, low-molecular compounds or mixtures thereof are applied on the substrate with the structured electrode. Examples of polymers are polyaniline, poly(p-phenylene-vinylene) and poly(2-methoxy-5-(2'-ethyl)-hexyloxy-p-phenylene-vinylene). Examples of low-molecular compounds that preferably transport positive charge carriers are N,N'-bis-(3-methylphenyl)-N,N'-bis-(phenyl)-benzidine (m-TPD), 4,4',4''-tris-(N-3-methylphenyl-N-phenyl-amino)-triphenylamine (m-MTDATA) and 4,4',4''-tris-(carbazole-9-yl)-triphenylamine (TCTA). Hydroxy-chinoline aluminum-III salt (Alq₃) that can be doped with suitable chromophores (chinacridone derivatives, aromatic hydrocarbons, etc.), for example, is employed ^{as} ~~as~~ emitter. As warranted, additional substances that influence the electro-optical and the long-term properties, such as copper phthalocyanine, can be present. The application of polymers usually ensues from the liquid phase with doctor blades or spin-coating; low-molecular and

oligomeric compounds are usually deposited from the vapor phase by vapor deposition or "physical vapor deposition" (PVD). The overall layer thickness can amount to between 10 nm and 10 ^{nm} ~~μm~~ it typically lies in the range between 50 and 200 nm.

5 A cooperating electrode (top electrode, cathode), which is usually composed of a metal, of a metal alloy or of a thin insulator layer and a thick metal layer, is applied onto the organic layer or layers. The manufacture of the cathode layer usually ensues with vapor phase deposition by means of thermal evaporation, electron beam evaporation or sputtering.

10 When metals are employed as cathode material, then these must have a low work function (typically < 3.7 eV) so that electrons can be efficiently injected into the organic semiconductor. Alkaline metals, alkaline earth metals or rare earth metals are usually employed for this purpose ^{and} the layer thickness lies between 0.2 nm and a few hundred nanometers but generally at a few 10 nanometers. Since, however, these
15 non-precious metals tend toward corrosion under atmospheric conditions, it is necessary to additionally apply a layer of a more precious, inert metal such as aluminum (Al), copper (Cu), silver (Ag) or gold (Au) onto the cathode layer that protects the non-precious metal layer against moisture and atmospheric oxygen.

For increasing the stability of the cathodes against a corrosion-caused hole
20 formation, an alloy composed of an efficiently electron-injecting but corrosion-susceptible non-precious metal (work function < 3.7 eV) and a corrosion-resistant ^{or} precious metal, such as Al, Cu, Ag and Au, is often employed instead of an unalloyed non-precious metal. The proportion of the non-precious metal in the alloy can amount to between a few tenths of a percent and approximately 90%. The alloys are usually
25 generated by simultaneous deposition of the metals from the vapor phase, for example by co-vapor deposition, simultaneous sputtering with a plurality of sources and sputtering upon employment of alloy targets. However, a layer of ^aprecious metal ~~or corrosion-resistant metal~~ such as Al, Cu, Ag or Au, is usually also additionally applied onto such cathodes as protection against corrosion.

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Cathodes composed of precious metals, i.e. metals having a work function > 3.7 eV, are very inefficient electron injectors when they are utilized in direct contact with the organic semiconductor. When, however, a thin insulating intermediate layer (layer thickness generally between 0.2 and 5 nm) is arranged between the uppermost, 5 electron-conducting organic layer and the metal electrode, then the efficiency of the light-emitting diodes rises substantially. Oxides such as aluminum oxide, alkaline and alkaline earth oxides and other oxides as well as alkaline and alkaline earth fluorides come into consideration as ^{the} insulating material for such an intermediate layer (in this respect, see Appl. Phys. Lett., Vol. 71 (1997), pages 2560 through 2562; 10 United States Letters Patent 5,677,572; European Published Application 0 822 603). A metal electrode that is composed of a pure metal or of a metal alloy is then applied onto the thin, insulating intermediate layer. The insulating material can thereby also be applied together with the electrode material by means of co-vapor deposition (Appl. Phys. Lett., Vol. 73 (1998), pages 1185 through 1187).

15 *Summary of the Invention*
An object of the invention is to fashion an organic electroluminescent component, particularly an organic light-emitting diode, such that, on the one hand, a hermetic seal of the top electrode can be foregone and, on the other hand, the selection of materials employable at the cathode side is greater.

This is inventively achieved by a component that is characterized by *it comprises*

20 -- a transparent bottom electrode situated on a substrate;
 -- a top electrode composed of a metal that is inert to oxygen and moisture;
 -- at least one organic function layer arranged between the bottom electrode and the top electrode; and
 -- a charge carrier injection layer containing a complex metal salt of the composition $(Me1)(Me2)F_{m+n}$, whereby the following applies:
 25 m and n are respectively a whole number corresponding to the valence of the metals Me1 and Me2 (the metal Me1 thereby has the valence m, the metal Me2 the valence n),
~~Me1 is selected from a group consisting of~~
 Me1 ~~is~~ Li, Na, K, Mg ~~or~~ Ca, and Ca

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is selected from a group consisting of

Me2 ~~*~~ Mg, Al, Ca, Zn, Ag, Sb, Ba, Sm ^{and} Yb,

with the prescription: Me1 \neq Me2.

The critical feature of the organic electroluminescent component of the invention is thus ~~comprised~~ in a specific structure at the cathode side, namely in the combination of a top electrode that is indifferent with respect to environmental influences with a charge carrier injection layer composed of a specific complex metal salt having the composition (Me1) (Me2)F_{m+n}, i.e. a double fluoride. As a result of this structure, a hermetic seal or, respectively, sealing of the top electrode can be omitted. As a result of the specific material for the charge carrier injection layer, not only is the offering for the materials employable at the cathode side broadened, this material also achieves an improvement of the emission properties, *which are* expressed in a clearly higher light yield, a reduced operating voltage and a longer service life during operation.

The charge carrier injection layer (composed of a specific complex metal salt) is preferably arranged as a thin insulating ~~layer~~ *either* between the top electrode and the organic function layer *or* between the uppermost function layer and the top electrode given the presence of a plurality of function layers. When an electron transport layer is also additionally located on the (uppermost) function layer given the component of the invention, then the charge carrier injection layer is arranged between this *transport* layer and the top electrode. In all of these instances, the thickness of the charge carrier injection layer preferably amounts to approximately 0.1 through 20 nm.

However, the charge carrier injection layer can also be quasi-integrated into the top electrode, into the (uppermost) organic function layer or into an electron transport layer that is potentially present, i.e. the complex metal salt is then a constituent part of one of *these* ~~said~~ layers. The production of such layers can advantageously ensue by means of co-vapor deposition of the corresponding materials, for example by co-vapor deposition of the top electrode material and of the complex metal salt.

The complex metal salt exhibits the composition $(Me1)(Me2)F_{m+n}$,
whereby m and n correspond to the valence of the respective metal. m = 1 (Li, Na, K)
or m = 2 (Mg, Ca) is valid for Me1; n = 1 (Ag) or n = 2 (Mg, Ca, Zn, Ba) or n = 3 (Al,
Sb, Sm, Yb) is valid for Me2. The metal Me1 is preferably lithium (Li); the metal
5 Me2 is preferably magnesium (Mg), aluminum (Al), calcium (Ca), silver (Ag) or
Barium (Ba).

Advantageously, one of the double fluorides $LiAgF_2$, $LiBaF_3$ and $LiAlF_4$ is
employed as ^{the} complex metal salt. More such double fluorides are, for example,
 $NaAgF_2$, $KAgF_2$, $LiMgF_3$, $LiCaF_3$, $CaAgF_3$ and $MgBaF_4$. Complex salts of this type
10 as well as methods for manufacturing them are known in and of themselves (in this
respect, see the exemplary embodiments as well as, for example, "Gmelins Handbuch
der Anorganischen Chemie", 8th Edition (1926), System Number 5 (fluorine), pages
58 through 72).

The top electrode, which generally comprises a thickness > 100 nm, is
15 preferably composed of one of the following metals: aluminum (Al), silver (Ag),
platinum (Pt) and gold (Au). The electrode material, however, can also be an alloy of
two of these metals. Magnesium (Mg), calcium (Ca), zinc (Zn), antimony (Sb) and
barium (Ba) come into consideration as ^{other} further metals for the top electrode.

The bottom electrode is generally composed of indium tin oxide (ITO).
20 ^{Other} Further possible materials for the bottom electrode are tin oxide and bismuth oxide.
Glass generally serves as ^{the} substrate for the bottom electrode.

The component of the invention preferably comprises two organic
function layers, namely an apertured conducting layer arranged at the bottom electrode
that transports positive charge carriers and an emission layer situated thereon that is
25 also referred to as ^{the} luminescence layer. Two or more apertured conducting layers can
also be advantageously utilized instead of one apertured conducting layer.

The materials for ^{these} said layers are known in and of themselves. In the
present case, N,N'-bis-(3-methylphenyl)-N,N'-bis(phenyl)-benzidine (m-TPD),
4,4',4''-tris-(N-1-naphthyl-N-phenylamino)-triphenylamine (naphdata) or N,N'-bis-
30 phenyl-N,N'-bis- α -naphthyl-benzidine (α -NPD) is preferably employed for the

apertured conducting layer or layers. The material for the emission layer is preferably hydroxyquinoline aluminum-III salt (Alq_3). Simultaneously, this material can also serve for the electron transport. For example, chinacridone can also be utilized for the emission layer, ^{and} one of the oxadiazole derivatives known for this purpose for a potentially present electron transport layer.

The invention offers the following, ^{additional} further advantages, particularly in view of organic light-emitting diodes:

-- Facilitated Handling

Due to the stability of the material of the top electrode, work need not be carried out under an inert gas atmosphere in the manufacture and further-processing of OLEDs.

-- Performance

Compared to top electrodes of non-precious metals, the operating voltage is clearly lowered and the light yield and efficiency are considerably enhanced.

-- Improved Properties

Compared, for example, to LiF as ^{the} material for the intermediate layer, compounds such as LiAlF_4 have the advantage that they are less hygroscopic, which facilitates the handling and storage. The double fluorides are also easier to evaporate and are less basic ^{than} LiF , as a result whereof the compatibility with the organic function layers is increased.

The invention shall be explained in still greater detail on the basis of exemplary embodiments and Figures.

25 Shown are: *Brief Description of the Drawings*

Figure 1 ^{is a diagrammatic view of} a traditional OLED display;

Figure 2 ^{is a diagrammatic view of} an OLED display of the invention;

Figure 3 ^{is a graph showing} luminance/voltage characteristics;

Figure 4 ^{is a graph showing} efficiency/luminance characteristics; ^{and}

30 Figure 5 ^{is a graph showing} a comparison of the luminance of various materials.

Description of the preferred embodiments

Example 1

Production of lithium aluminum fluoride LiAlF_4

Lithium aluminum hydride LiAlH_4 is carefully hydrolyzed with distilled water ^{and} conversion is subsequently undertaken with hydrofluoric acid (HF) ^{being} in excess.

- 5 The complex metal salt LiAlF_4 that thereby precipitates out is extracted, repeatedly washed with water and ethanol and then dried.

Example 2

Production of lithium silver fluoride LiAgF_2

- 10 A solution of stoichiometric quantities of lithium hydroxide and silver acetate is glacial acetic acid is converted with hydrofluoric acid (HF) ^{being} in excess upon exclusion of light ^{and} the complex metal salt LiAgF_2 thereby precipitates out. The complex salt is extracted after the addition of the same volume of ethanol, is washed with ethanol and dried.

Example 3

- 15 Production of lithium barium fluoride LiBa_3

An aqueous solution of stoichiometric quantities of lithium hydroxide and barium hydroxide is converted with hydrofluoric acid (HF) ^{being} in excess ^{and}. The complex metal salt LiBaF_3 precipitates out when chilled (cooling with ice); it is extracted, repeatedly washed with ethanol and then dried.

- 20 The complex metal salt LiCaF_3 is produced in a corresponding way, whereby the reaction solution is constricted as warranted.

The complex metal salt LiMgF_3 can be produced in the same way ^{and} lithium methylate and magnesium methylate are then utilized as ^{the} initial substances.

Example 4

- 25 Manufacture of a traditional OLED display ¹⁰ (10) with a Mg/Ag cathode (see Figure 1)

An ITO layer ¹²(~~12~~) having a thickness of approximately 100 nm is applied onto a glass substrate ¹¹(~~11~~). This layer is then photolithographically structured in such a way that a stripe-shaped structure ^{13 produced}arises. A layer ¹³of m-TPD (~~13~~) having a thickness of approximately 100 nm is first applied by thermal evaporation onto the coated substrate pre-treated in this way, followed by a layer ¹⁴(~~14~~) of Alq₃ having a thickness of approximately 65 nm.

A layer ¹⁵(~~15~~) of a magnesium-silver alloy (Mg:Ag mixing ratio 10:1) having a thickness of approximately 150 nm is applied onto the organic layer ¹⁴(~~14~~) by thermal evaporation with two simultaneously operated evaporator sources, and a layer ¹⁶(~~16~~) of pure silver having a thickness of approximately 150 nm is applied on said ~~the~~ layer ¹⁵(~~15~~), likewise by thermal evaporation. The metal layers are thereby vapor-deposited through a mask with stripe-shaped openings, so that cathode stripes that lie perpendicular to the ITO stripes ^{are produced}arise. Organic light-emitting diodes with an active area of 2 x 2 mm² respectively ^{are produced}arise in this way at the intersections of the ITO tracks with the metal tracks -- together with the organic layers lying therebetween. During operation, the ITO layer is positively contacted ^{and}the metal tracks are negatively contacted.

Example 5

Manufacture of an OLED display ²⁰(~~20~~) of the invention (see Figure 2)

An ITO layer ²²(~~22~~) having a thickness of approximately 100 nm is applied onto a glass substrate ²¹(~~21~~). This layer is then photolithographically structured in such a way that a stripe-shaped structure ^{is produced}arises. A layer ²³of m-TPD (~~23~~) having a thickness of approximately 100 nm is first applied by thermal evaporation onto the coated substrate pre-treated in this way, followed by a layer ²⁴(~~24~~) of Alq₃ having a thickness of approximately 65 nm.

A layer ²⁵(~~25~~) of LiAlF₄ having a thickness of approximately 1 nm is applied by thermal evaporation onto the ~~organic~~ layer ²⁴(~~24~~), and a layer ²⁶(~~26~~) of aluminum -- serving as ^atop electrode -- having a thickness of approximately 150 nm is applied onto said layer ²⁵(~~25~~), likewise by thermal evaporation. The two layers are

thereby vapor-deposited through a mask with stripe-shaped openings, corresponding to Example 4, so that organic light-emitting diodes ^{are produced} arise. During operation, the ITO layer is positively contacted, ^{and} the top electrode ^{is} negatively contacted.

The results of measurements at the OLEDs corresponding to Examples 4 and 5 are compiled in Table 1. The threshold voltage (of the electroluminescence), the voltage and the efficiency (respectively given a luminance of 1500 cd/m²), the maximum luminance and the luminance given a current density of 50 mA/cm²) are thereby recited as characteristic data.

Table 1

10

| Example | Threshold voltage [V] | Voltage [V] at 1500 cd/m ² | Efficiency [lm/W] at 1500 cd/m ² | Maximum luminance [cd/m ²] | Luminance [cd/m ²] at 50 mA/cm ² |
|---------|-----------------------|---------------------------------------|---|--|---|
| 4 | 2.08 | 14.48 | 0.677 | 15957 | 1544 |
| 5 | 1.87 | 14.12 | 0.720 | 18801 | 1605 |

It can be seen that the threshold voltage and the operating voltage of the display of the invention (Example 5) lie below the corresponding values given the traditional display (Example 4), even though the thickness of the LiAlF₄ was not optimized. The values for the efficiency and the luminances that are achieved given the display of the invention lie above the corresponding values of the traditional display.

Figure 3 shows the luminance/voltage characteristics of the displays according to Examples 4 and 5. The increased luminance of the display of the invention can be clearly seen from this illustration.

The following can be stated overall:

- The display of the invention (Example 5) employs a cathode of aluminum with which efficiencies are normally achieved that lie approximately 40 to 50% below the corresponding values given Mg/Ag cathodes (Example 4). Aluminum, on the other hand, is more stable than magnesium vis a vis environmental influences such as atmospheric oxygen and moisture.

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-- Due to the introduction of a thin LiAlF_4 between the organic function layers and the Al cathode, however, the efficiency of OLEDs having an Al cathode can be increased, namely even above the corresponding values of OLEDs with Mg/Ag cathode. In this way, high-efficiency OLEDs with stable cathode can be constructed.

Example 6

Manufacture of an OLED display with a Mg/Ag cathode

An ITO layer having a thickness of approximately 100 nm is applied onto a glass substrate. This layer is then photolithographically structured in such a way that a stripe-shaped structure ^{is produced,} arises. A layer of naphdata having a thickness of approximately 55 nm is first applied by thermal evaporation onto the coated substrate pre-treated in this way, followed by a layer of α -NPD having a thickness of approximately 5 nm, and, finally, a layer of Alq_3 having a thickness of approximately 65 nm.

15 A layer of a magnesium-silver alloy (Mg:Ag mixing ratio 10:1) having a thickness of approximately 150 nm is applied onto the uppermost organic layer (of Alq_3) by thermal evaporation with two simultaneously operated evaporator sources, and a layer of pure silver having a thickness of approximately 150 nm is applied on ^{the} said uppermost organic layer, likewise by thermal evaporation. The metal layers are thereby vapor-deposited through a mask with stripe-shaped openings, so that cathode stripes that lie perpendicular to the ITO stripes ^{are produced.} Organic light-emitting diodes with an active area of $2 \times 2 \text{ mm}^2$ respectively ^{are produced} arise in this way at the intersections of the ITO tracks with the metal tracks -- together with the organic layers lying therebetween. During operation, the ITO layer is positively contacted ^{and} the metal tracks are negatively contacted.

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Example 7

Manufacture of an OLED display with an Al cathode

Corresponding to Example 6, a display having three organic function layers is constructed. A layer of aluminum having a thickness of 150 nm is then applied in a corresponding way by thermal evaporation onto the uppermost organic layer (of Alq₃).

Example 8

Manufacture of an OLED display with an Al cathode and an LiF intermediate layer

A display with three organic function layers is constructed corresponding to Example 6. A layer of LiF having a thickness of approximately 0.5 nm is then applied onto the uppermost organic layer (of Alq₃) by thermal evaporation, and a layer of aluminum having a thickness of approximately 150 nm is applied on said uppermost organic layer. The two layers are thereby vapor-deposited through a mask having stripe-shaped openings in conformity with Example 6, so that organic light-emitting diodes ^{are produced} arise. During operation, the ITO layer is positively contacted, the Al cathode ^{is} negatively contacted.

Example 9

Manufacture of an OLED display with an Al cathode and a LiAlF₄ charge carrier injection layer

A display with three organic function layers is constructed corresponding to Example 8. A layer of LiAlF₄ having a thickness of approximately 0.5 nm is then applied by thermal evaporation onto the uppermost organic layer (of Alq₃), and a layer of aluminum -- serving as top electrode -- having a thickness of approximately 150 nm is applied onto ^{the} said LiAlF₄ layer, likewise by thermal evaporation. The structuring and the contacting ensue in conformity with Example 8.

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Example 10

Manufacture of an OLED display with an Al cathode and a LiAgF_2 charge carrier injection layer

5 A display with three organic function layers is constructed corresponding to Example 6. A layer of LiAgF_2 having a thickness of approximately 0.5 nm is then applied by thermal evaporation onto the uppermost organic layer (of Alq_3), and a layer of aluminum -- serving as^a top electrode -- having a thickness of approximately 150 nm is applied onto said LiAlF_4 layer, likewise by thermal evaporation. The structuring and the contacting ensue in conformity with Example 8.

10 Example 11

Manufacture of an OLED display with an Al cathode and a LiBaF_3 charge carrier injection layer

15 A display with three organic function layers is constructed corresponding to Example 6. A layer of LiBaF_3 having a thickness of approximately 0.5 nm is then applied by thermal evaporation onto the uppermost organic layer (of Alq_3), and a layer of aluminum -- serving as^a top electrode -- having a thickness of approximately 150 nm is applied onto ~~said~~^{the} LiAlF_4 layer, likewise by thermal evaporation. The structuring and the contacting ensue in conformity with Example 8.

20 The results of measurements at the OLEDs corresponding to Examples 6 through 11 are compiled in Table 2. The threshold voltage (of the electroluminescence), the voltage and the efficiency (respectively given a luminance of 1500 cd/m^2), the maximum luminance and the luminance given a current density of 50 mA/cm^2) are thereby recited as characteristic data.

Table 2

| Example | Threshold voltage [V] | Voltage [V] at 1500 cd/m ² | Efficiency [lm/W] at 1500 cd/m ² | Luminance [cd/m ²] at 50 mA/cm ² |
|---------|-----------------------|---------------------------------------|---|---|
| 6 | 3.19 | 9.96 | 1.08 | 1722 |
| 7 | 7.15 | 16.52 | 0.48 | 1275 |
| 8 | 3.17 | 9.47 | 1.19 | 1809 |
| 9 | 4.23 | 11.97 | 0.88 | 1684 |
| 10 | 3.49 | 10.86 | 1.00 | 1745 |
| 11 | 2.56 | 9.58 | 1.26 | 1948 |

It can be seen that the threshold voltages and the operating voltages of the displays of the invention (Examples 9 through 11) that comprise an Al cathode and a charge carrier injection layer composed of a complex metal salt are comparable to the values that are obtained given displays with a Mg/Ag cathode or, respectively, with an Al cathode and a LiF intermediate layer (Examples 6 and 8) and lie clearly below the corresponding values given a display with a pure Al cathode (Example 7). The displays of the invention are also comparable to the Mg/Ag and Al-LiF displays in view of the efficiency and the luminance, whereby a display with a LiBaF₃ charge carrier injection layer (Example 11), in particular, exhibits high values.

Figure 4 shows the efficiency/luminance characteristics of the Examples 6 through 11. In particular, the superior position of an Al-LiBaF₃ of the invention can be clearly seen from this illustration.

It can be stated overall that the efficiency of LEDs with an Al cathode can be boosted above the corresponding values of OLEDs with a Mg/Ag cathode by introducing thin layers of a complex metal salt such as LiAlF₄, LiAgF₂ and LiBaF₃ between the organic function layers and the cathode. High-efficiency OLEDs with stable contact can be constructed in this way.

The values for the luminance (given a current density of 50 mA/cm²) of the materials according to Examples 6 through 11 are compared to one another in Figure 5. The good results that can be achieved with the displays of the invention also *can be derived* therefrom.

We claim:
Patent Claims

1. Organic electroluminescent component, particularly an organic light-emitting diode, characterized by
 - a transparent bottom electrode (22) situated on a substrate (21);
 - 5 -- a top electrode (26) composed of a metal that is inert to oxygen and moisture;
 - at least one organic function layer (23, 24) arranged between the bottom electrode (22) and the top electrode (26); and
 - a charge carrier injection layer (25) containing a complex metal salt of the composition (Me1) (Me2) F_{m+n} , whereby the following applies:
 10 m and n are respectively a whole number corresponding to the valence of the metals Me1 and Me2 (the metal Me1 thereby has the valence m, the metal Me2 the valence n),
 Me1 = Li, Na, K, Mg or Ca,
 15 Me2 = Mg, Al, Ca, Zn, Ag, Sb, Ba, Sm or Yb,
 with the prescription: Me1 \neq Me2.
2. Component according to claim 1, characterized in that the top electrode (26) is composed of aluminum, silver, platinum or gold or of an alloy of two of these metals.
- 20 3. Component according to claim 1 or 2, characterized in that the charge carrier injection layer (25) is arranged between the top electrode (26) and the organic function layer (24).
4. Component according to one of the claims 1 through 3, characterized in that the charge carrier injection layer (25) comprises a thickness between 0.1 and 20
 25 nm.
5. Component according to one or more of the claims 1 through 4, characterized in that the metal Me1 is lithium (Li) and/or the metal Me2 is magnesium (Mg), aluminum (Al), calcium (Ca), silver (Ag) or barium (Ba).
6. Component according to claim 5, characterized in that the complex
 30 metal salt is LiAlF₄, LiAgF₂ or LiBaF₃.

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Abstract of the Disclosure

Organic Electroluminescent Component

The organic electroluminescent component ^{is an} of the invention comprises the following constituents:—

- 5 -- a transparent bottom electrode (22) situated on a substrate (21);
- a top electrode (26) composed of a metal that is inert to oxygen and moisture;
- at least one organic function layer (23, 24) arranged between the bottom electrode (22) and the top electrode (26); and
- 10 -- a charge carrier injection layer (25) containing a complex metal salt of the composition (Me1) (Me2) F_{m+n} , whereby the following applies:
m and n are respectively a whole number corresponding to the valence of the metals Me1 and Me2 (the metal Me1 thereby has the valence m, the metal Me2 the valence n),
15 Me1 ~~is~~ ^{is selected from a group consisting of} Li, Na, K, Mg ~~or~~ Ca, and
Me2 ~~is~~ ^{is selected from a group consisting of} Mg, Al, Ca, Zn, Ag, Sb, Ba, Sm ~~or~~ Yb, ^{and}
with the prescription: Me1 \neq Me2.

~~Figure 2~~

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TITLE

"ORGANIC ELECTROLUMINESCENT COMPONENT"

BACKGROUND OF THE INVENTION

5 The invention is directed to an organic electroluminescent component, particularly an organic light-emitting diode.

10 The visualization of data is constantly increasing in significance due to the great increase in the amount of information. The technology of flat picture screens ("flat panel displays") was developed therefor for employment in mobile and portable electronic devices. The market of flat panel displays is currently largely dominated by the technology of liquid crystal displays (LC displays). In addition to cost-beneficial manufacture, low electrical power consumption, low weight and slight space requirement, however, the technology of LC displays also exhibits serious disadvantages.

15 LC displays are not self-emitting and can therefore only be easily read or recognized given especially beneficial ambient light conditions. This makes a back-illumination device necessary in most instances, but this multiplies the thickness of the flat panel display. Moreover, the majority part of the electrical power consumption of the display is then needed for the illumination, and a higher voltage is required for the operation of the lamps or fluorescent tubes, which higher voltage is usually generated from batteries or accumulators with the assistance of "voltage-up converters". Other disadvantages are the highly limited observation angles of LC displays and the long switching times of individual pixels, which switching times typically lie at a few milliseconds and also are highly temperature-dependent. The delayed image build-up is considered extremely disturbing, for example, given
25 utilization in means of conveyance or given video applications.

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There are other flat panel display technologies in addition to LC displays, for example the technology of flat display panel cathode ray tubes, of vacuum-fluorescence displays and of inorganic thin-film electroluminescent displays. However, either these technologies have not yet achieved the required degree of technological maturity or -- due to high operating voltages or, respectively, high manufacturing costs -- they are only conditionally suited for utilization in portable electronic devices.

Displays on the basis of organic light-emitting diodes, which are called OLEDs, do not exhibit these disadvantages. The necessity of a back-illumination is eliminated due to the self-emissivity, as a result whereof the space requirement and the electrical power consumption are considerably reduced. The switching times lie at about one microsecond and are only slightly temperature-dependent, which enables employment for video applications. The reading angle amounts to nearly 180°, and polarization films that are required given LC displays are eliminated, so that a greater brightness of the display elements can be achieved. Further advantages are the employability of flexible and non-planar substrates as well as a simple and cost-beneficial manufacture.

The construction of organic light-emitting diodes typically ensues in the following way.

A transparent substrate, for example glass, is coated with a transparent electrode (bottom electrode, anode), composed, for example, of indium tin oxide (ITO). Dependent on the application, the transparent electrode is then structured with the assistance of a photolithographic process.

One or more organic layers composed of polymers, oligomers, low-molecular compounds or mixtures thereof are applied on the substrate with the structured electrode. Examples of polymers are polyaniline, poly(p-phenylene-

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vinylene) and poly(2-methoxy-5-(2'-ethyl)-hexyloxy-p-phenylene-vinylene). Examples of low-molecular compounds that preferably transport positive charge carriers are N,N'-bis-(3-methylphenyl)-N,N'-bis-(phenyl)-benzidine (m-TPD), 4,4',4''-tris-(N-3-methylphenyl-N-phenyl-amino)-triphenylamine (m-MTDATA) and 4,4',4''-tris-(carbazole-9-yl)-triphenylamine (TCTA). Hydroxy-chinoline aluminum-III salt (Alq_3) that can be doped with suitable chromophores (chinacridone derivatives, aromatic hydrocarbons, etc.), for example, is employed as an emitter. As warranted, additional substances that influence the electro-optical and the long-term properties, such as copper phthalocyanine, can be present. The application of polymers usually ensues from the liquid phase with doctor blades or spin-coating; low-molecular and oligomeric compounds are usually deposited from the vapor phase by vapor deposition or "physical vapor deposition" (PVD). The overall layer thickness can amount to between 10 nm and 10 μm and it typically lies in the range between 50 and 200 nm.

A cooperating electrode (top electrode, cathode), which is usually composed of a metal, of a metal alloy or of a thin insulator layer and a thick metal layer, is applied onto the organic layer or layers. The manufacture of the cathode layer usually ensues with vapor phase deposition by means of thermal evaporation, electron beam evaporation or sputtering.

When metals are employed as cathode material, then these must have a low work function (typically < 3.7 eV) so that electrons can be efficiently injected into the organic semiconductor. Alkaline metals, alkaline earth metals or rare earth metals are usually employed for this purpose and the layer thickness lies between 0.2 nm and a few hundred nanometers but generally at a few 10 nanometers. Since, however, these non-precious metals tend toward corrosion under atmospheric conditions, it is necessary to additionally apply a layer of a more precious, inert metal

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such as aluminum (Al), copper (Cu), silver (Ag) or gold (Au) onto the cathode layer that protects the non-precious metal layer against moisture and atmospheric oxygen.

For increasing the stability of the cathodes against a corrosion-caused hole formation, an alloy composed of an efficiently electron-injecting but corrosion-susceptible non-precious metal (work function < 3.7 eV) and a corrosion-resistant or precious metal, such as Al, Cu, Ag and Au, is often employed instead of an unalloyed non-precious metal. The proportion of the non-precious metal in the alloy can amount to between a few tenths of a percent and approximately 90%. The alloys are usually generated by simultaneous deposition of the metals from the vapor phase, for example by co-vapor deposition, simultaneous sputtering with a plurality of sources and sputtering upon employment of alloy targets. However, a layer of a precious metal or corrosion-resistant metal, such as Al, Cu, Ag or Au, is usually also additionally applied onto such cathodes as protection against corrosion.

Cathodes composed of precious metals, i.e. metals having a work function > 3.7 eV, are very inefficient electron injectors when they are utilized in direct contact with the organic semiconductor. When, however, a thin insulating intermediate layer (layer thickness generally between 0.2 and 5 nm) is arranged between the uppermost, electron-conducting organic layer and the metal electrode, then the efficiency of the light-emitting diodes rises substantially. Oxides such as aluminum oxide, alkaline and alkaline earth oxides and other oxides as well as alkaline and alkaline earth fluorides come into consideration as the insulating material for such an intermediate layer (in this respect, see Appl. Phys. Lett., Vol. 71 (1997), pages 2560 through 2562; United States Letters Patent 5,677,572; European Published Application 0 822 603). A metal electrode that is composed of a pure metal or of a metal alloy is then applied onto the thin, insulating intermediate layer. The insulating material can thereby also be applied together with the electrode

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material by means of co-vapor deposition (Appl. Phys. Lett., Vol. 73 (1998), pages 1185 through 1187).

SUMMARY OF THE INVENTION

5 An object of the invention is to fashion an organic electroluminescent component, particularly an organic light-emitting diode, such that, on the one hand, a hermetic seal of the top electrode can be foregone and, on the other hand, the selection of materials employable at the cathode side is greater.

This is inventively achieved by a component that is characterized by or comprises

- 10 -- a transparent bottom electrode situated on a substrate;
- a top electrode composed of a metal that is inert to oxygen and moisture;
- at least one organic function layer arranged between the bottom electrode and the top electrode; and
- a charge carrier injection layer containing a complex metal salt of the
- 15 composition $(Me1)(Me2)F_{m+n}$, whereby the following applies:
- m and n are respectively a whole number corresponding to the valence of the metals Me1 and Me2 (the metal Me1 thereby has the valence m, the metal Me2 the valence n),
- Me1 is selected from a group consisting of Li, Na, K, Mg and Ca.
- 20 Me2 is selected from a group consisting of Mg, Al, Ca, Zn, Ag, Sb, Ba, Sm and Yb,
- with the prescription: $Me1 \neq Me2$.

The critical feature of the organic electroluminescent component of the invention is thus in a specific structure at the cathode side, namely in the combination

25 of a top electrode that is indifferent with respect to environmental influences with a

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charge carrier injection layer composed of a specific complex metal salt having the composition $(Me1)(Me2)F_{m+n}$, i.e. a double fluoride. As a result of this structure, a hermetic seal or, respectively, sealing of the top electrode can be omitted. As a result of the specific material for the charge carrier injection layer, not only is the offering for the materials employable at the cathode side broadened, this material also achieves an improvement of the emission properties, which are expressed in a clearly higher light yield, a reduced operating voltage and a longer service life during operation.

The charge carrier injection layer (composed of a specific complex metal salt) is preferably arranged as a thin insulating layer either between the top electrode and the organic function layer or between the uppermost function layer and the top electrode given the presence of a plurality of function layers. When an electron transport layer is also additionally located on the (uppermost) function layer given the component of the invention, then the charge carrier injection layer is arranged between this transport layer and the top electrode. In all of these instances, the thickness of the charge carrier injection layer preferably amounts to approximately 0.1 through 20 nm.

However, the charge carrier injection layer can also be quasi-integrated into the top electrode, into the (uppermost) organic function layer or into an electron transport layer that is potentially present, i.e. the complex metal salt is then a constituent part of one of these layers. The production of such layers can advantageously ensue by means of co-vapor deposition of the corresponding materials, for example by co-vapor deposition of the top electrode material and of the complex metal salt.

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The complex metal salt exhibits the composition $(\text{Me1})(\text{Me2})\text{F}_{m+n}$, whereby m and n correspond to the valence of the respective metal. $m = 1$ (Li, Na, K) or $m = 2$ (Mg, Ca) is valid for Me1; $n = 1$ (Ag) or $n = 2$ (Mg, Ca, Zn, Ba) or $n = 3$ (Al, Sb, Sm, Yb) is valid for Me2. The metal Me1 is preferably lithium (Li); the metal Me2 is preferably magnesium (Mg), aluminum (Al), calcium (Ca), silver (Ag) or Barium (Ba).

Advantageously, one of the double fluorides LiAgF_2 , LiBaF_3 and LiAlF_4 is employed as the complex metal salt. More such double fluorides are, for example, NaAgF_2 , KAgF_2 , LiMgF_3 , LiCaF_3 , CaAgF_3 and MgBaF_4 . Complex salts of this type as well as methods for manufacturing them are known in and of themselves (in this respect, see the exemplary embodiments as well as, for example, "Gmelins Handbuch der Anorganischen Chemie", 8th Edition (1926), System Number 5 (fluorine), pages 58 through 72).

The top electrode, which generally comprises a thickness > 100 nm, is preferably composed of one of the following metals: aluminum (Al), silver (Ag), platinum (Pt) and gold (Au). The electrode material, however, can also be an alloy of two of these metals. Magnesium (Mg), calcium (Ca), zinc (Zn), antimony (Sb) and barium (Ba) come into consideration as other metals for the top electrode.

The bottom electrode is generally composed of indium tin oxide (ITO). Other possible materials for the bottom electrode are tin oxide and bismuth oxide. Glass generally serves as the substrate for the bottom electrode.

The component of the invention preferably comprises two organic function layers, namely an apertured conducting layer arranged at the bottom electrode that transports positive charge carriers and an emission layer situated thereon that is also referred to as the luminescence layer. Two or more apertured conducting layers can also be advantageously utilized instead of one apertured conducting layer.

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The materials for these layers are known in and of themselves. In the present case, N,N'-bis-(3-methylphenyl)-N,N'-bis(phenyl)-benzidine (m-TPD), 4,4',4''-tris-(N-1-naphthyl-N-phenylamino)-triphenylamine (naphdata) or N,N'-bis-phenyl-N,N'-bis- α -naphthyl-benzidine (α -NPD) is preferably employed for the apertured conducting layer or layers. The material for the emission layer is preferably hydroxyquinoline aluminum-III salt (Alq₃). Simultaneously, this material can also serve for the electron transport. For example, chinacridone can also be utilized for the emission layer, and one of the oxadiazole derivatives known for this purpose for a potentially present electron transport layer.

The invention offers the following, additional advantages, particularly in view of organic light-emitting diodes:

-- Facilitated Handling

Due to the stability of the material of the top electrode, work need not be carried out under an inert gas atmosphere in the manufacture and further-processing of OLEDs.

-- Performance

Compared to top electrodes of non-precious metals, the operating voltage is clearly lowered and the light yield and efficiency are considerably enhanced.

-- Improved Properties

Compared, for example, to LiF as the material for the intermediate layer, compounds such as LiAlF₄ have the advantage that they are less hygroscopic, which facilitates the handling and storage. The double fluorides are also easier to evaporate and are less basic than LiF, as a result whereof the compatibility with the organic function layers is increased.

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The invention shall be explained in still greater detail on the basis of exemplary embodiments and Figures.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a diagrammatic view of a traditional OLED display;

Figure 2 is a diagrammatic view of an OLED display of the invention;

Figure 3 is a graph showing luminance/voltage characteristics;

Figure 4 is a graph showing efficiency/luminance characteristics;

Figure 5 is a graph showing a comparison of the luminance of various materials.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

Example 1

Production of lithium aluminum fluoride LiAlF_4

Lithium aluminum hydride LiAlH_4 is carefully hydrolyzed with distilled water and conversion is subsequently undertaken with hydrofluoric acid (HF) being in excess. The complex metal salt LiAlF_4 that thereby precipitates out is extracted, repeatedly washed with water and ethanol and then dried.

Example 2

Production of lithium silver fluoride LiAgF_2

A solution of stoichiometric quantities of lithium hydroxide and silver acetate in glacial acetic acid is converted with hydrofluoric acid (HF) being in excess upon exclusion of light and the complex metal salt LiAgF_2 thereby precipitates out. The complex salt is extracted after the addition of the same volume of ethanol, is washed with ethanol and dried.

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Example 3

Production of lithium barium fluoride LiBa_3F_3

An aqueous solution of stoichiometric quantities of lithium hydroxide and barium hydroxide is converted with hydrofluoric acid (HF) being in excess. The
5 complex metal salt LiBaF_3 precipitates out when chilled (cooling with ice) and it is extracted, repeatedly washed with ethanol and then dried.

The complex metal salt LiCaF_3 is produced in a corresponding way, whereby the reaction solution is constricted as warranted.

The complex metal salt LiMgF_3 can be produced in the same way and
10 lithium methyllate and magnesium methyllate are then utilized as the initial substances.

Example 4

Manufacture of a traditional OLED display 10 with a Mg/Ag cathode (see Figure 1)

An ITO layer 12 having a thickness of approximately 100 nm is applied
15 onto a glass substrate 11. This layer is then photolithographically structured in such a way that a stripe-shaped structure is produced. A layer 13 of m-TPD having a thickness of approximately 100 nm is first applied by thermal evaporation onto the coated substrate pre-treated in this way, followed by a layer 14 of Alq_3 having a thickness of approximately 65 nm.

20 A layer 15 of a magnesium-silver alloy (Mg:Ag mixing ratio 10:1) having a thickness of approximately 150 nm is applied onto the organic layer 14 by thermal evaporation with two simultaneously operated evaporator sources, and a layer 16 of pure silver having a thickness of approximately 150 nm is applied on the layer 15, likewise by thermal evaporation. The metal layers are thereby vapor-deposited
25 through a mask with stripe-shaped openings, so that cathode stripes that lie

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perpendicular to the ITO stripes are produced. Organic light-emitting diodes with an active area of $2 \times 2 \text{ mm}^2$ respectively are produced in this way at the intersections of the ITO tracks with the metal tracks -- together with the organic layers lying therebetween. During operation, the ITO layer is positively contacted and the metal tracks are negatively contacted.

Example 5

Manufacture of an OLED display 20 of the invention (see Figure 2)

An ITO layer 22 having a thickness of approximately 100 nm is applied onto a glass substrate 21. This layer is then photolithographically structured in such a way that a stripe-shaped structure is produced. A layer 23 of m-TPD having a thickness of approximately 100 nm is first applied by thermal evaporation onto the coated substrate pre-treated in this way, followed by a layer 24 of Alq_3 having a thickness of approximately 65 nm.

A layer 25 of LiAlF_4 having a thickness of approximately 1 nm is applied by thermal evaporation onto the layer 24, and a layer 26 of aluminum -- serving as a top electrode -- having a thickness of approximately 150 nm is applied onto said layer 25, likewise by thermal evaporation. The two layers are thereby vapor-deposited through a mask with stripe-shaped openings, corresponding to Example 4, so that organic light-emitting diodes are produced. During operation, the ITO layer is positively contacted and the top electrode is negatively contacted.

The results of measurements at the OLEDs corresponding to Examples 4 and 5 are compiled in Table 1. The threshold voltage (of the electroluminescence), the voltage and the efficiency (respectively given a luminance of 1500 cd/m^2), the maximum luminance and the luminance given a current density of 50 mA/cm^2) are thereby recited as characteristic data.

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Table 1

| Exempl e | Threshold voltage [V] | Voltage [V] at 1500 cd/m ² | Efficiency [lm/W] at 1500 cd/m ² | Maximum luminance [cd/m ²] | Luminance [cd/m ²] at 50 mA/cm ² |
|-------------|--------------------------|--|---|--|---|
| 4 | 2.08 | 14.48 | 0.677 | 15957 | 1544 |
| 5 | 1.87 | 14.12 | 0.720 | 18801 | 1605 |

It can be seen that the threshold voltage and the operating voltage of the display of the invention (Example 5) lie below the corresponding values given the traditional display (Example 4), even though the thickness of the LiAlF₄ was not optimized. The values for the efficiency and the luminances that are achieved given the display of the invention lie above the corresponding values of the traditional display.

Figure 3 shows the luminance/voltage characteristics of the displays according to Examples 4 and 5. The increased luminance of the display of the invention can be clearly seen from this illustration.

The following can be stated overall:

- The display of the invention (Example 5) employs a cathode of aluminum with which efficiencies are normally achieved that lie approximately 40 to 50% below the corresponding values given Mg/Ag cathodes (Example 4). Aluminum, on the other hand, is more stable than magnesium vis a vis environmental influences such as atmospheric oxygen and moisture.
- Due to the introduction of a thin LiAlF₄ between the organic function layers and the Al cathode, however, the efficiency of OLEDs having an Al cathode can be increased, namely even above the corresponding values of OLEDs with Mg/Ag cathode. In this way, high-efficiency OLEDs with stable cathode can be constructed.

Example 6

Manufacture of an OLED display with a Mg/Ag cathode

An ITO layer having a thickness of approximately 100 nm is applied onto a glass substrate. This layer is then photolithographically structured in such a way that a stripe-shaped structure is produced. A layer of naphdata having a thickness of approximately 55 nm is first applied by thermal evaporation onto the coated substrate pre-treated in this way, followed by a layer of α -NPD having a thickness of approximately 5 nm, and, finally, a layer of Alq_3 having a thickness of approximately 65 nm.

A layer of a magnesium-silver alloy (Mg:Ag mixing ratio 10:1) having a thickness of approximately 150 nm is applied onto the uppermost organic layer (of Alq_3) by thermal evaporation with two simultaneously operated evaporator sources, and a layer of pure silver having a thickness of approximately 150 nm is applied on the uppermost organic layer, likewise by thermal evaporation. The metal layers are thereby vapor-deposited through a mask with stripe-shaped openings, so that cathode stripes that lie perpendicular to the ITO stripes are produced. Organic light-emitting diodes with an active area of $2 \times 2 \text{ mm}^2$ respectively are produced in this way at the intersections of the ITO tracks with the metal tracks -- together with the organic layers lying therebetween. During operation, the ITO layer is positively contacted and the metal tracks are negatively contacted.

Example 7

Manufacture of an OLED display with an Al cathode

Corresponding to Example 6, a display having three organic function layers is constructed. A layer of aluminum having a thickness of 150 nm is then applied in

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a corresponding way by thermal evaporation onto the uppermost organic layer (of Alq_3).

Example 8

Manufacture of an OLED display with an Al cathode and an LiF intermediate layer

5 A display with three organic function layers is constructed corresponding to Example 6. A layer of LiF having a thickness of approximately 0.5 nm is then applied onto the uppermost organic layer (of Alq_3) by thermal evaporation, and a layer of aluminum having a thickness of approximately 150 nm is applied on the uppermost organic layer. The two layers are thereby vapor-deposited through a mask
10 having stripe-shaped openings in conformity with Example 6, so that organic light-emitting diodes are produced. During operation, the ITO layer is positively contacted and the Al cathode is negatively contacted.

Example 9

Manufacture of an OLED display with an Al cathode and a LiAlF_4 charge carrier
15 injection layer

 A display with three organic function layers is constructed corresponding to Example 8. A layer of LiAlF_4 having a thickness of approximately 0.5 nm is then applied by thermal evaporation onto the uppermost organic layer (of Alq_3), and a layer of aluminum -- serving as top electrode -- having a thickness of approximately
20 150 nm is applied onto the LiAlF_4 layer, likewise by thermal evaporation. The structuring and the contacting ensue in conformity with Example 8.

Example 10

Manufacture of an OLED display with an Al cathode and a LiAgF_2 charge carrier injection layer

5 A display with three organic function layers is constructed corresponding to Example 6. A layer of LiAgF_2 having a thickness of approximately 0.5 nm is then applied by thermal evaporation onto the uppermost organic layer (of Alq_3), and a layer of aluminum -- serving as a top electrode -- having a thickness of approximately 150 nm is applied onto said LiAlF_4 layer, likewise by thermal evaporation. The structuring and the contacting ensue in conformity with Example 8.

10 Example 11

Manufacture of an OLED display with an Al cathode and a LiBaF_3 charge carrier injection layer

15 A display with three organic function layers is constructed corresponding to Example 6. A layer of LiBaF_3 having a thickness of approximately 0.5 nm is then applied by thermal evaporation onto the uppermost organic layer (of Alq_3), and a layer of aluminum -- serving as a top electrode -- having a thickness of approximately 150 nm is applied onto the LiAlF_4 layer, likewise by thermal evaporation. The structuring and the contacting ensue in conformity with Example 8.

20 The results of measurements at the OLEDs corresponding to Examples 6 through 11 are compiled in Table 2. The threshold voltage (of the electroluminescence), the voltage and the efficiency (respectively given a luminance of 1500 cd/m^2), the maximum luminance and the luminance given a current density of 50 mA/cm^2) are thereby recited as characteristic data.

Table 2

| Example | Threshold voltage [V] | Voltage [V] at 1500 cd/m ² | Efficiency [lm/W] at 1500 cd/m ² | Luminance [cd/m ²] at 50 mA/cm ² |
|---------|-----------------------|---------------------------------------|---|---|
| 6 | 3.19 | 9.96 | 1.08 | 1722 |
| 7 | 7.15 | 16.52 | 0.48 | 1275 |
| 8 | 3.17 | 9.47 | 1.19 | 1809 |
| 9 | 4.23 | 11.97 | 0.88 | 1684 |
| 10 | 3.49 | 10.86 | 1.00 | 1745 |
| 11 | 2.56 | 9.58 | 1.26 | 1948 |

It can be seen that the threshold voltages and the operating voltages of the displays of the invention (Examples 9 through 11) that comprise an Al cathode and a charge carrier injection layer composed of a complex metal salt are comparable to the values that are obtained given displays with a Mg/Ag cathode or, respectively, with an Al cathode and a LiF intermediate layer (Examples 6 and 8) and lie clearly below the corresponding values given a display with a pure Al cathode (Example 7). The displays of the invention are also comparable to the Mg/Ag and Al-LiF displays in view of the efficiency and the luminance, whereby a display with a LiBaF₃ charge carrier injection layer (Example 11), in particular, exhibits high values.

Figure 4 shows the efficiency/luminance characteristics of the Examples 6 through 11. In particular, the superior position of an Al-LiBaF₃ of the invention can be clearly seen from this illustration.

It can be stated overall that the efficiency of LEDs with an Al cathode can be boosted above the corresponding values of OLEDs with a Mg/Ag cathode by introducing thin layers of a complex metal salt such as LiAlF₄, LiAgF₂ and LiBaF₃ between the organic function layers and the cathode. High-efficiency OLEDs with stable contact can be constructed in this way.

The values for the luminance (given a current density of 50 mA/cm²) of the materials according to Examples 6 through 11 are compared to one another in Figure 5. The good results that can be achieved with the displays of the invention also can be derived therefrom.

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ORGANIC ELECTROLUMINESCENT COMPONENT

The invention is directed to an organic electroluminescent component, particularly an organic light-emitting diode.

The visualization of data is constantly increasing in significance due to the great increase in the amount of information. The technology of flat picture screens ("flat panel displays") was developed therefor for employment in mobile and portable electronic devices. The market of flat panel displays is currently largely dominated by the technology of liquid crystal displays (LC displays). In addition to cost-beneficial manufacture, low electrical power consumption, low weight and slight space requirement, however, the technology of LC displays also exhibits serious disadvantages.

LC displays are not self-emitting and can therefore only be easily read or recognized given especially beneficial ambient light conditions. This makes a back-illumination device necessary in most instances, but this multiplies the thickness of the flat panel display. Moreover, the majority part of the electrical power consumption is then needed for the illumination, and a higher voltage is required for the operation of the lamps or fluorescent tubes; this is usually generated from batteries or accumulators with the assistance of "voltage-up converters". Other disadvantages are the highly limited observation angles of LC displays and the long switching times of individual pixels, these typically lying at a few milliseconds and also being highly temperature-dependent. The delayed image build-up is considered extremely disturbing, for example, given utilization in means of conveyance or given video applications.

There are other flat panel display technologies in addition to LC displays, for example the technology of flat display panel cathode ray tubes, of vacuum-fluorescence displays and of inorganic thin-film electroluminescent displays. These technologies, however, have either not yet achieved the required degree of technological maturity or -- due to high operating voltages or, respectively, high

manufacturing costs -- they are only conditionally suited for utilization in portable electronic devices.

Displays on the basis of organic light-emitting diodes (=OLEDs) do not exhibit said disadvantages. The necessity of a back-illumination is eliminated due to the self-emissivity, as a result whereof the space requirement and the electrical power consumption are considerably reduced. The switching times lie at about one microsecond and are only slightly temperature-dependent, which enables employment for video applications. The reading angle amounts to nearly 180°, and polarization films as required given LC displays are eliminated, so that a greater brightness of the display elements can be achieved. Further advantages are the employability of flexible and non-planar substrates as well as a simple and cost-beneficial manufacture.

The construction of organic light-emitting diodes typically ensues in the following way.

A transparent substrate, for example glass, is coated with a transparent electrode (bottom electrode, anode), composed, for example, of indium tin oxide (ITO). Dependent on the application, the transparent electrode is then structured with the assistance of a photolithographic process.

One or more organic layers composed of polymers, oligomers, low-molecular compounds or mixtures thereof are applied on the substrate with the structured electrode. Examples of polymers are polyaniline, poly(p-phenylene-vinylene) and poly(2-methoxy-5-(2'-ethyl)-hexyloxy-p-phenylene-vinylene). Examples of low-molecular compounds that preferably transport positive charge carriers are N,N'-bis-(3-methylphenyl)-N,N'-bis-(phenyl)-benzidine (m-TPD), 4,4',4''-tris-(N-3-methylphenyl-N-phenyl-amino)-triphenylamine (m-MTDATA) and 4,4',4''-tris-(carbazole-9-yl)-triphenylamine (TCTA). Hydroxy-chinoline aluminum-III salt (Alq₃) that can be doped with suitable chromophores (chinacridone derivatives, aromatic hydrocarbons, etc.), for example, is employed as emitter. As warranted, additional substances that influence the electro-optical and the long-term properties, such as copper phthalocyanine, can be present. The application of polymers usually ensues from the liquid phase with doctor blades or spin-coating; low-molecular and

oligomeric compounds are usually deposited from the vapor phase by vapor deposition or “physical vapor deposition” (PVD). The overall layer thickness can amount to between 10 nm and 10 μm ; it typically lies in the range between 50 and 200 nm.

5 A cooperating electrode (top electrode, cathode), which is usually composed of a metal, of a metal alloy or of a thin insulator layer and a thick metal layer, is applied onto the organic layer or layers. The manufacture of the cathode layer usually ensues with vapor phase deposition by means of thermal evaporation, electron beam evaporation or sputtering.

10 When metals are employed as cathode material, then these must have a low work function (typically < 3.7 eV) so that electrons can be efficiently injected into the organic semiconductor. Alkaline metals, alkaline earth metals or rare earth metals are usually employed for this purpose; the layer thickness lies between 0.2 nm and a few hundred nanometers but generally at a few 10 nanometers. Since, however, these
15 non-precious metals tend toward corrosion under atmospheric conditions, it is necessary to additionally apply a layer of a more precious, inert metal such as aluminum (Al), copper (Cu), silver (Ag) or gold (Au) onto the cathode layer that protects the non-precious metal layer against moisture and atmospheric oxygen.

For increasing the stability of the cathodes against a corrosion-caused hole
20 formation, an alloy composed of an efficiently electron-injecting but corrosion-susceptible non-precious metal (work function < 3.7 eV) and a corrosion-resistant precious metal, such as Al, Cu, Ag and Au, is often employed instead of an unalloyed non-precious metal. The proportion of the non-precious metal in the alloy can amount to between a few tenths of a percent and approximately 90%. The alloys are usually
25 generated by simultaneous deposition of the metals from the vapor phase, for example by co-vapor deposition, simultaneous sputtering with a plurality of sources and sputtering upon employment of alloy targets. However, a layer of precious metal, such as Al, Cu, Ag or Au, is usually also additionally applied onto such cathodes as protection against corrosion.

Cathodes composed of precious metals, i.e. metals having a work function > 3.7 eV, are very inefficient electron injectors when they are utilized in direct contact with the organic semiconductor. When, however, a thin insulating intermediate layer (layer thickness generally between 0.2 and 5 nm) is arranged between the uppermost, electron-conducting organic layer and the metal electrode, then the efficiency of the light-emitting diodes rises substantially. Oxides such as aluminum oxide, alkaline and alkaline earth oxides and other oxides as well as alkaline and alkaline earth fluorides come into consideration as insulating material for such an intermediate layer (in this respect, see Appl. Phys. Lett., Vol. 71 (1997), pages 2560 through 2562; United States Letters Patent 5,677,572; European Published Application 0 822 603). A metal electrode that is composed of a pure metal or of a metal alloy is then applied onto the thin, insulating intermediate layer. The insulating material can thereby also be applied together with the electrode material by means of co-vapor deposition (Appl. Phys. Lett., Vol. 73 (1998), pages 1185 through 1187).

An object of the invention is to fashion an organic electroluminescent component, particularly an organic light-emitting diode, such that, on the one hand, a hermetic seal of the top electrode can be foregone and, on the other hand, the selection of materials employable at the cathode side is greater.

This is inventively achieved by a component that is characterized by

- a transparent bottom electrode situated on a substrate;
- a top electrode composed of a metal that is inert to oxygen and moisture;
- at least one organic function layer arranged between the bottom electrode and the top electrode; and
- a charge carrier injection layer containing a complex metal salt of the composition $(Me1)(Me2)F_{m+n}$, whereby the following applies:
 m and n are respectively a whole number corresponding to the valence of the metals Me1 and Me2 (the metal Me1 thereby has the valence m , the metal Me2 the valence n),
 $Me1 = Li, Na, K, Mg$ or Ca ,

Me2 = Mg, Al, Ca, Zn, Ag, Sb, Ba, Sm or Yb,

with the prescription: Me1 \neq Me2.

The critical feature of the organic electroluminescent component of the invention is thus comprised in a specific structure at the cathode side, namely in the combination of a top electrode that is indifferent with respect to environmental influences with a charge carrier injection layer composed of a specific complex metal salt having the composition (Me1) (Me2)F_{m+n}, i.e. a double fluoride. As a result of this structure, a hermetic seal or, respectively, sealing of the top electrode can be omitted. As a result of the specific material for the charge carrier injection layer, not only is the offering for the materials employable at the cathode side broadened, this material also achieves an improvement of the emission properties, expressed in a clearly higher light yield, a reduced operating voltage and a longer service life during operation.

The charge carrier injection layer (composed of a specific complex metal salt) is preferably arranged as a thin insulating layer between the top electrode and the organic function layer, between the uppermost function layer and the top electrode given the presence of a plurality of function layers. When an electron transport layer is also additionally located on the (uppermost) function layer given the component of the invention, then the charge carrier injection layer is arranged between this layer and the top electrode. In all of these instances, the thickness of the charge carrier injection layer preferably amounts to approximately 0.1 through 20 nm.

However, the charge carrier injection layer can also be quasi-integrated into the top electrode, into the (uppermost) organic function layer or into an electron transport layer that is potentially present, i.e. the complex metal salt is then a constituent part of one of said layers. The production of such layers can advantageously ensue by means of co-vapor deposition of the corresponding materials, for example by co-vapor deposition of the top electrode material and of the complex metal salt.

The complex metal salt exhibits the composition (Me1) (Me2) F_{m+n} , whereby m and n correspond to the valence of the respective metal. m = 1 (Li, Na, K) or m = 2 (Mg, Ca) is valid for Me1; n = 1 (Ag) or n = 2 (Mg, Ca, Zn, Ba) or n = 3 (Al, Sb, Sm, Yb) is valid for Me2. The metal Me1 is preferably lithium (Li); the metal
 5 Me2 is preferably magnesium (Mg), aluminum (Al), calcium (Ca), silver (Ag) or Barium (Ba).

Advantageously, one of the double fluorides LiAgF₂, LiBaF₃ and LiAlF₄ is employed as complex metal salt. More such double fluorides are, for example, NaAgF₂, KAgF₂, LiMgF₃, LiCaF₃, CaAgF₃ and MgBaF₄. Complex salts of this type
 10 as well as methods for manufacturing them are known in and of themselves (in this respect, see the exemplary embodiments as well as, for example, "Gmelins Handbuch der Anorganischen Chemie", 8th Edition (1926), System Number 5 (fluorine), pages 58 through 72).

The top electrode, which generally comprises a thickness > 100 nm, is
 15 preferably composed of one of the following metals: aluminum (Al), silver (Ag), platinum (Pt) and gold (Au). The electrode material, however, can also be an alloy of two of these metals. Magnesium (Mg), calcium (Ca), zinc (Zn), antimony (Sb) and barium (Ba) come into consideration as further metals for the top electrode.

The bottom electrode is generally composed of indium tin oxide (ITO).
 20 Further possible materials for the bottom electrode are tin oxide and bismuth oxide. Glass generally serves as substrate for the bottom electrode.

The component of the invention preferably comprises two organic function layers, namely an apertured conducting layer arranged at the bottom electrode that transports positive charge carriers and an emission layer situated thereon that is
 25 also referred to as luminescence layer. Two or more apertured conducting layers can also be advantageously utilized instead of one apertured conducting layer.

The materials for said layers are known in and of themselves. In the present case, N,N'-bis-(3-methylphenyl)-N,N'-bis(phenyl)-benzidine (m-TPD), 4,4',4''-tris-(N-1-naphthyl-N-phenylamino)-triphenylamine (naphdata) or N,N'-bis-
 30 phenyl-N,N'-bis- α -naphthyl-benzidine (α -NPD) is preferably employed for the

apertured conducting layer or layers. The material for the emission layer is preferably hydroxyquinoline aluminum-III salt (Alq_3). Simultaneously, this material can also serve for the electron transport. For example, chinacridone can also be utilized for the emission layer, one of the oxadiazole derivatives known for this purpose for a potentially present electron transport layer.

The invention offers the following, further advantages, particularly in view of organic light-emitting diodes:

-- Facilitated Handling

Due to the stability of the material of the top electrode, work need not be carried out under an inert gas atmosphere in the manufacture and further-processing of OLEDs.

-- Performance

Compared to top electrodes of non-precious metals, the operating voltage is clearly lowered and the light yield and efficiency are considerably enhanced.

-- Improved Properties

Compared, for example, to LiF as material for the intermediate layer, compounds such as LiAlF_4 have the advantage that they are less hygroscopic, which facilitates the handling and storage. The double fluorides are also easier to evaporate and are less basic (than LiF), as a result whereof the compatibility with the organic function layers is increased.

The invention shall be explained in still greater detail on the basis of exemplary embodiments and Figures.

Shown are:

- Figure 1 a traditional OLED display;
- Figure 2 an OLED display of the invention;
- Figure 3 luminance/voltage characteristics;
- Figure 4 efficiency/luminance characteristics;
- Figure 5 a comparison of the luminance of various materials.

Example 1

Production of lithium aluminum fluoride LiAlF_4

Lithium aluminum hydride LiAlH_4 is carefully hydrolyzed with distilled water; conversion is subsequently undertaken with hydrofluoric acid (HF) in excess.

- 5 The complex metal salt LiAlF_4 that thereby precipitates out is extracted, repeatedly washed with water and ethanol and then dried.

Example 2

Production of lithium silver fluoride LiAgF_2

- 10 A solution of stoichiometric quantities of lithium hydroxide and silver acetate is glacial acetic acid is converted with hydrofluoric acid (HF) in excess upon exclusion of light; the complex metal salt LiAgF_2 thereby precipitates out. The complex salt is extracted after the addition of the same volume of ethanol, is washed with ethanol and dried.

Example 3

- 15 Production of lithium barium fluoride LiBa_3

An aqueous solution of stoichiometric quantities of lithium hydroxide and barium hydroxide is converted with hydrofluoric acid (HF) in excess. The complex metal salt LiBaF_3 precipitates out when chilled (cooling with ice); it is extracted, repeatedly washed with ethanol and then dried.

- 20 The complex metal salt LiCaF_3 is produced in a corresponding way, whereby the reaction solution is constricted as warranted.

The complex metal salt LiMgF_3 can be produced in the same way; lithium methyllate and magnesium methyllate are then utilized as initial substances.

Example 4

- 25 Manufacture of a traditional OLED display (10) with a Mg/Ag cathode (see Figure 1)

An ITO layer (12) having a thickness of approximately 100 nm is applied onto a glass substrate (11). This layer is then photolithographically structured in such a way that a stripe-shaped structure arises. A layer of m-TPD (13) having a thickness of approximately 100 nm is first applied by thermal evaporation onto the coated substrate pre-treated in this way, followed by a layer (14) of Alq₃ having a thickness of approximately 65 nm.

A layer (15) of a magnesium-silver alloy (Mg:Ag mixing ratio 10:1) having a thickness of approximately 150 nm is applied onto the organic layer (14) by thermal evaporation with two simultaneously operated evaporator sources, and a layer (16) of pure silver having a thickness of approximately 150 nm is applied on said layer (15), likewise by thermal evaporation. The metal layers are thereby vapor-deposited through a mask with stripe-shaped openings, so that cathode stripes that lie perpendicular to the ITO stripes arise. Organic light-emitting diodes with an active area of 2 x 2 mm² respectively arise in this way at the intersections of the ITO tracks with the metal tracks -- together with the organic layers lying therebetween. During operation, the ITO layer is positively contacted; the metal tracks are negatively contacted.

Example 5

Manufacture of an OLED display (20) of the invention (see Figure 2)

An ITO layer (22) having a thickness of approximately 100 nm is applied onto a glass substrate (21). This layer is then photolithographically structured in such a way that a stripe-shaped structure arises. A layer of m-TPD (23) having a thickness of approximately 100 nm is first applied by thermal evaporation onto the coated substrate pre-treated in this way, followed by a layer (24) of Alq₃ having a thickness of approximately 65 nm.

A layer (25) of LiAlF₄ having a thickness of approximately 1 nm is applied by thermal evaporation onto the organic layer (24), and a layer (26) of aluminum -- serving as top electrode -- having a thickness of approximately 150 nm is applied onto said layer (25), likewise by thermal evaporation. The two layers are

thereby vapor-deposited through a mask with stripe-shaped openings, corresponding to Example 4, so that organic light-emitting diodes arise. During operation, the ITO layer is positively contacted, the top electrode negatively.

The results of measurements at the OLEDs corresponding to Examples 4 and 5 are compiled in Table 1. The threshold voltage (of the electroluminescence), the voltage and the efficiency (respectively given a luminance of 1500 cd/m²), the maximum luminance and the luminance given a current density of 50 mA/cm² are thereby recited as characteristic data.

Table 1

| Example | Threshold voltage [V] | Voltage [V] at 1500 cd/m ² | Efficiency [lm/W] at 1500 cd/m ² | Maximum luminance [cd/m ²] | Luminance [cd/m ²] at 50 mA/cm ² |
|---------|-----------------------|---------------------------------------|---|--|---|
| 4 | 2.08 | 14.48 | 0.677 | 15957 | 1544 |
| 5 | 1.87 | 14.12 | 0.720 | 18801 | 1605 |

It can be seen that the threshold voltage and the operating voltage of the display of the invention (Example 5) lie below the corresponding values given the traditional display (Example 4), even though the thickness of the LiAlF₄ was not optimized. The values for the efficiency and the luminances that are achieved given the display of the invention lie above the corresponding values of the traditional display.

Figure 3 shows the luminance/voltage characteristics of the displays according to Examples 4 and 5. The increased luminance of the display of the invention can be clearly seen from this illustration.

The following can be stated overall:

- The display of the invention (Example 5) employs a cathode of aluminum with which efficiencies are normally achieved that lie approximately 40 to 50% below the corresponding values given Mg/Ag cathodes (Example 4). Aluminum, on the other hand, is more stable than magnesium vis a vis environmental influences such as atmospheric oxygen and moisture.

-- Due to the introduction of a thin LiAlF_4 between the organic function layers and the Al cathode, however, the efficiency of OLEDs having an Al cathode can be increased, namely even above the corresponding values of OLEDs with Mg/Ag cathode. In this way, high-efficiency OLEDs with stable cathode can be constructed.

Example 6

Manufacture of an OLED display with a Mg/Ag cathode

An ITO layer having a thickness of approximately 100 nm is applied onto a glass substrate. This layer is then photolithographically structured in such a way that a stripe-shaped structure arises. A layer of naphdata having a thickness of approximately 55 nm is first applied by thermal evaporation onto the coated substrate pre-treated in this way, followed by a layer of α -NPD having a thickness of approximately 5 nm, and, finally, a layer of Alq_3 having a thickness of approximately 65 nm.

A layer of a magnesium-silver alloy (Mg:Ag mixing ratio 10:1) having a thickness of approximately 150 nm is applied onto the uppermost organic layer (of Alq_3) by thermal evaporation with two simultaneously operated evaporator sources, and a layer of pure silver having a thickness of approximately 150 nm is applied on said uppermost organic layer, likewise by thermal evaporation. The metal layers are thereby vapor-deposited through a mask with stripe-shaped openings, so that cathode stripes that lie perpendicular to the ITO stripes arise. Organic light-emitting diodes with an active area of $2 \times 2 \text{ mm}^2$ respectively arise in this way at the intersections of the ITO tracks with the metal tracks -- together with the organic layers lying therebetween. During operation, the ITO layer is positively contacted; the metal tracks are negatively contacted.

Example 7

Manufacture of an OLED display with an Al cathode

Corresponding to Example 6, a display having three organic function layers is constructed. A layer of aluminum having a thickness of 150 nm is then applied in a corresponding way by thermal evaporation onto the uppermost organic layer (of Alq₃).

Example 8

Manufacture of an OLED display with an Al cathode and an LiF intermediate layer

A display with three organic function layers is constructed corresponding to Example 6. A layer of LiF having a thickness of approximately 0.5 nm is then applied onto the uppermost organic layer (of Alq₃) by thermal evaporation, and a layer of aluminum having a thickness of approximately 150 nm is applied on said uppermost organic layer. The two layers are thereby vapor-deposited through a mask having stripe-shaped openings in conformity with Example 6, so that organic light-emitting diodes arise. During operation, the ITO layer is positively contacted, the Al cathode negatively.

Example 9

Manufacture of an OLED display with an Al cathode and a LiAlF₄ charge carrier injection layer

A display with three organic function layers is constructed corresponding to Example 8. A layer of LiAlF₄ having a thickness of approximately 0.5 nm is then applied by thermal evaporation onto the uppermost organic layer (of Alq₃), and a layer of aluminum -- serving as top electrode -- having a thickness of approximately 150 nm is applied onto said LiAlF₄ layer, likewise by thermal evaporation. The structuring and the contacting ensue in conformity with Example 8.

Example 10

Manufacture of an OLED display with an Al cathode and a LiAgF_2 charge carrier injection layer

- 5 A display with three organic function layers is constructed corresponding to Example 6. A layer of LiAgF_2 having a thickness of approximately 0.5 nm is then applied by thermal evaporation onto the uppermost organic layer (of Alq_3), and a layer of aluminum -- serving as top electrode -- having a thickness of approximately 150 nm is applied onto said LiAlF_4 layer, likewise by thermal evaporation. The structuring and the contacting ensue in conformity with Example 8.

10 Example 11

Manufacture of an OLED display with an Al cathode and a LiBaF_3 charge carrier injection layer

- 15 A display with three organic function layers is constructed corresponding to Example 6. A layer of LiBaF_3 having a thickness of approximately 0.5 nm is then applied by thermal evaporation onto the uppermost organic layer (of Alq_3), and a layer of aluminum -- serving as top electrode -- having a thickness of approximately 150 nm is applied onto said LiAlF_4 layer, likewise by thermal evaporation. The structuring and the contacting ensue in conformity with Example 8.

- 20 The results of measurements at the OLEDs corresponding to Examples 6 through 11 are compiled in Table 2. The threshold voltage (of the electroluminescence), the voltage and the efficiency (respectively given a luminance of 1500 cd/m^2), the maximum luminance and the luminance given a current density of 50 mA/cm^2) are thereby recited as characteristic data.

Table 2

| Example | Threshold voltage [V] | Voltage [V] at 1500 cd/m ² | Efficiency [lm/W] at 1500 cd/m ² | Luminance [cd/m ²] at 50 mA/cm ² |
|---------|-----------------------|---------------------------------------|---|---|
| 6 | 3.19 | 9.96 | 1.08 | 1722 |
| 7 | 7.15 | 16.52 | 0.48 | 1275 |
| 8 | 3.17 | 9.47 | 1.19 | 1809 |
| 9 | 4.23 | 11.97 | 0.88 | 1684 |
| 10 | 3.49 | 10.86 | 1.00 | 1745 |
| 11 | 2.56 | 9.58 | 1.26 | 1948 |

It can be seen that the threshold voltages and the operating voltages of the displays of the invention (Examples 9 through 11) that comprise an Al cathode and a charge carrier injection layer composed of a complex metal salt are comparable to the values that are obtained given displays with a Mg/Ag cathode or, respectively, with an Al cathode and a LiF intermediate layer (Examples 6 and 8) and lie clearly below the corresponding values given a display with a pure Al cathode (Example 7). The displays of the invention are also comparable to the Mg/Ag and Al-LiF displays in view of the efficiency and the luminance, whereby a display with a LiBaF₃ charge carrier injection layer (Example 11), in particular, exhibits high values.

Figure 4 shows the efficiency/luminance characteristics of the Examples 6 through 11. In particular, the superior position of an Al-LiBaF₃ of the invention can be clearly seen from this illustration.

It can be stated overall that the efficiency of LEDs with an Al cathode can be boosted above the corresponding values of OLEDs with a Mg/Ag cathode by introducing thin layers of a complex metal salt such as LiAlF₄, LiAgF₂ and LiBaF₃ between the organic function layers and the cathode. High-efficiency OLEDs with stable contact can be constructed in this way.

The values for the luminance (given a current density of 50 mA/cm²) of the materials according to Examples 6 through 11 are compared to one another in Figure 5. The good results that can be achieved with the displays of the invention also derive therefrom.

Patent Claims

1. Organic electroluminescent component, particularly an organic light-emitting diode, characterized by

- a transparent bottom electrode (22) situated on a substrate (21);
- 5 -- a top electrode (26) composed of a metal that is inert to oxygen and moisture;
- at least one organic function layer (23, 24) arranged between the bottom electrode (22) and the top electrode (26); and
- a charge carrier injection layer (25) containing a complex metal salt of the
- 10 composition $(Me1)(Me2)F_{m+n}$, whereby the following applies:
 m and n are respectively a whole number corresponding to the valence of the metals $Me1$ and $Me2$ (the metal $Me1$ thereby has the valence m , the metal $Me2$ the valence n),
 $Me1 = Li, Na, K, Mg$ or Ca ,
- 15 $Me2 = Mg, Al, Ca, Zn, Ag, Sb, Ba, Sm$ or Yb ,
 with the prescription: $Me1 \neq Me2$.

2. Component according to claim 1, characterized in that the top electrode (26) is composed of aluminum, silver, platinum or gold or of an alloy of two of these metals.

20 3. Component according to claim 1 or 2, characterized in that the charge carrier injection layer (25) is arranged between the top electrode (26) and the organic function layer (24).

4. Component according to one of the claims 1 through 3, characterized in that the charge carrier injection layer (25) comprises a thickness between 0.1 and 20

25 nm.

5. Component according to one or more of the claims 1 through 4, characterized in that the metal $Me1$ is lithium (Li) and/or the metal $Me2$ is magnesium (Mg), aluminum (Al), calcium (Ca), silver (Ag) or barium (Ba).

6. Component according to claim 5, characterized in that the complex

30 metal salt is $LiAlF_4$, $LiAgF_2$ or $LiBaF_3$.

7. Component according to one or more of the claims 1 through 6,
characterized in that two organic function layers (23, 24) are arranged between the
bottom electrode (22) and the top electrode (26), whereby an apertured conducting
layer (23) is located on the bottom electrode (22) and an emission layer (24) is located
5 on said conducting layer (23).

8. Component according to claim 7, characterized in that the apertured
conducting layer (23) contains N,N'-bis-(3-methylphenyl)-N,N'-bis(phenyl)-
benzidine, 4,4',4''-tris-(N-1-naphthyl-N-phenylamino)-triphenylamine or N,N'-bis-
phenyl-N,N'-bis- α -naphthyl-benzidine and/or the emission layer (24)
10 hydroxyquinoline aluminum-III salt.

9. Component according to one or more of the claims 1 through 8,
characterized in that the bottom electrode (22) is composed of indium tin oxide.

10. Component according to one or more of the claims 1 through 9,
characterized in that a electron transport layer is arranged on the at least one organic
15 function layer (23, 24).

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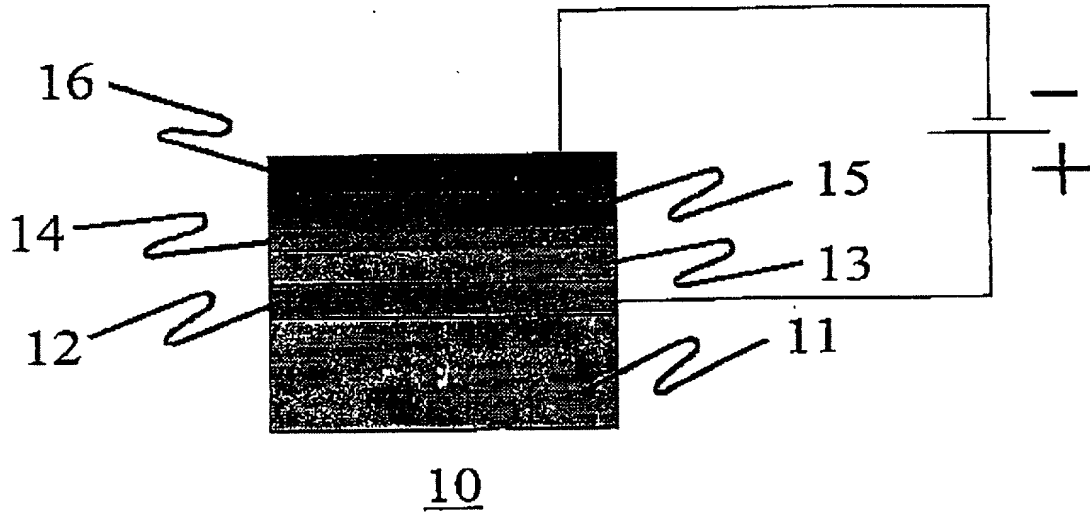


FIG 1

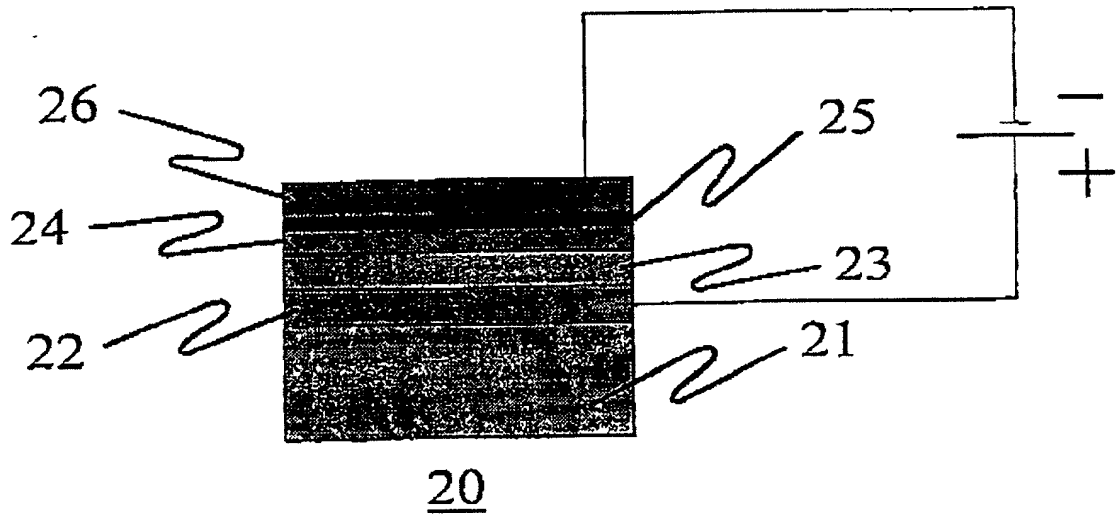


FIG 2

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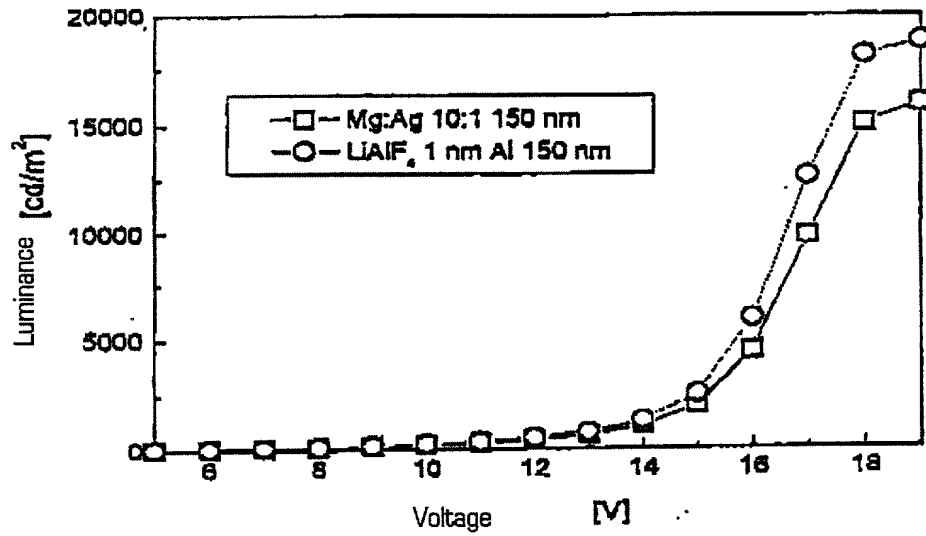


FIG 3

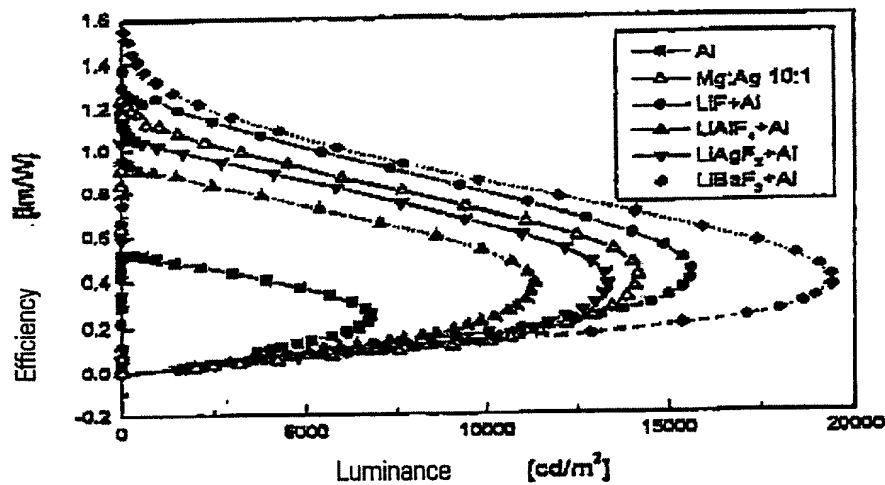


FIG 4

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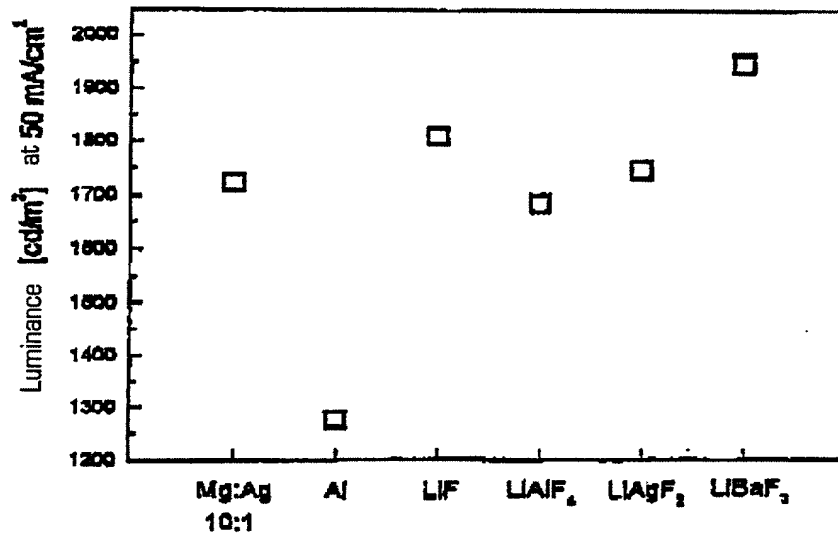


FIG 5

202070-06226650

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY

(Includes Reference to PCT International Applications)

ATTORNEY'S
DOCKET NUMBER
P01,0300

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

ORGANIC ELECTROLUMINESCENT COMPONENT

the specification of which (check only one item below):

- ☒ is attached hereto.
- ☐ was filed as United States application
Serial No. _____
on _____,
and was amended
on _____ (if applicable).
- ☒ was filed as PCT international application
Number PCT/DE00/00783
on March 13, 2000,
and was amended under PCT Article 19
on _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. 119:

| COUNTRY (if PCT indicate "PCT") | APPLICATION NUMBER | DATE OF FILING (day, month, year) | PRIORITY CLAIMED UNDER 35 USC 119 |
|------------------------------------|--------------------|--------------------------------------|---|
| Germany | 199 13 350.6 | March 24, 1999 | <input checked="" type="checkbox"/> YES <input type="checkbox"/> NO |
| | | | <input type="checkbox"/> YES <input type="checkbox"/> NO |
| | | | <input type="checkbox"/> YES <input type="checkbox"/> NO |
| | | | <input type="checkbox"/> YES <input type="checkbox"/> NO |
| | | | <input type="checkbox"/> YES <input type="checkbox"/> NO |

Combined Declaration For Patent Application and Power of Attorney
(Continued) (Includes Reference to PCT International Applications)

ATTORNEY'S DOCKET NO.
P01,0300

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56 which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application:

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| U.S. APPLICATIONS | | | STATUS (Check one) | | |
|---------------------------------------|------------------|---------------------------------------|--------------------|---------|-----------|
| U.S. APPLICATION NUMBER | U.S. FILING DATE | | PATENTED | PENDING | ABANDONED |
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| PCT APPLICATIONS DESIGNATING THE U.S. | | | | | |
| PCT APPLICATION NO | PCT FILING DATE | U.S. SERIAL NUMBERS ASSIGNED (if any) | | | |
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And I hereby appoint all Attorneys Identified by United States Patent & Trademark Office Customer Number 26574, who are all members of the firm of Schiff Hardin and Waite.

Telephone 312/258-5500 Patent Department

my attorneys with full power of substitution and revocation, to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

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| | POST OFFICE ADDRESS | POST OFFICE ADDRESS Nackendorf 27 | CITY D91315 Hoechst | STATE & ZIP CODE/COUNTRY Germany |
| 202 | FULL NAME OF INVENTOR | FAMILY NAME Stoessel | FIRST GIVEN NAME Matthias | SECOND GIVEN NAME |
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| 203 | FULL NAME OF INVENTOR | FAMILY NAME | FIRST GIVEN NAME | SECOND GIVEN NAME |
| | RESIDENCE & CITIZENSHIP | CITY | STATE OR FOREIGN COUNTRY | COUNTRY OF CITIZENSHIP |
| | POST OFFICE ADDRESS | POST OFFICE ADDRESS | CITY | STATE & ZIP CODE/COUNTRY |

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

| | | |
|---------------------------|---|---------------------------|
| SIGNATURE OF INVENTOR 201 | SIGNATURE OF INVENTOR 202 <i>Matthias Stoessel</i> | SIGNATURE OF INVENTOR 203 |
| DATE | DATE 10-10-01 | DATE |

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY

(Includes Reference to PCT International Applications)

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Combined Declaration For Patent Application and Power of Attorney
(Continued) (Includes Reference to PCT International Applications)

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| | | | | | |
| PCT APPLICATIONS DESIGNATING THE U.S. | | | | | |
| PCT APPLICATION NO | PCT FILING DATE | U.S. SERIAL NUMBERS ASSIGNED (if any) | | | |
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| | | | | | |

And I hereby appoint all Attorneys Identified by United States Patent & Trademark Office Customer Number 26574, who are all members of the firm of Schiff Hardin and Waite.
Telephone 312-258-5500 Patent Department
my attorneys with full power of substitution and revocation, to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

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| 201 | FULL NAME OF INVENTOR | FAMILY NAME <u>Kanitz</u> | FIRST GIVEN NAME <u>Andreas</u> | SECOND GIVEN NAME |
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| | POST OFFICE ADDRESS | POST OFFICE ADDRESS Nackendorf 27 | CITY D91315 Hoechst | STATE & ZIP CODE/COUNTRY Germany |
| 202 | FULL NAME OF INVENTOR | FAMILY NAME <u>Stoessel</u> | FIRST GIVEN NAME <u>Matthias</u> | SECOND GIVEN NAME |
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SIGNATURE OF INVENTOR 201

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DATE Oct 06 / 01 / 2001

DATE

DATE